PB82-189556

Suspended Particulate Matter
A Report to Congress

(U.S.) Environmental Protection Agency
Research Triangle Park, NC.

Feb 79

U.S. Department of Commerce
National Technical Information Service
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TECHNICAL REPORT DATA *(Please read Instructions on the reverse before completing)*		
1. REPORT NO. EPA-600/9-79-006	2.	3. RECIPIENT'S ACCESSION NO. PB82 189556
4. TITLE AND SUBTITLE SUSPENDED PARTICULATE MATTER: A Report to Congress		5. REPORT DATE February 1979 6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Lucile Adamson, Howard University Robert Bruce		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS U.S. Environmental Protection Agency Environmental Criteria Assessment Office (MD-52) Research Triangle Park, NC 27711		10. PROGRAM ELEMENT NO. 11. CONTRACT/GRANT NO. 1HA882
12. SPONSORING AGENCY NAME AND ADDRESS U.S. Environmental Protection Agency Office of Research and Development Office of Health and Environmental Assessment (RD-689) 401 M Street, SW, Washington, DC 20460		13. TYPE OF REPORT AND PERIOD COVERED Final 14. SPONSORING AGENCY CODE EPA/600/00
15. SUPPLEMENTARY NOTES		

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16. ABSTRACT

This report is in response to Section 403(a)(1) of the Clean Air Act as Amended August 1977. The report covers: 1) a review of the physical and chemical characteristics of airborne particles (source, composition, and sampling site as related to size); 2) a review of the effects of particulate matter on public welfare (ecological, materials, atmospheric, aesthetic); 3) the status of human exposure to airborne particles as related to source; and 4) a review of the effects of airborne particles on human health (lung deposition, chemical composition, interactions, and potentiating conditions).

Although there is a wide divergence of opinion among experts and scientific groups with respect to the issues of particulates (cf. Appendix A), the following can be concluded from the available information:

1. High levels of airborne particles have been associated with episodes of high pollution during the past, especially in the United Kingdom and the United States.

2. Although pollution levels have declined in many U.S. localities in recent decades, there is still need for improvement in several of our cities.

3. Additional research is needed to improve the scientific basis for future airborne particle standards as outlined by EPA (cf. Dr. Cortesi in the Culver Pilot Study of Particulate Matter).

17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
18. DISTRIBUTION STATEMENT DISTRIBUTE TO PUBLIC	19. SECURITY CLASS *(This Report)* UNCLASSIFIED 20. SECURITY CLASS *(This page)* UNCLASSIFIED	21. NO. OF PAGES 85 22. PRICE

PB82-189556

United States
Environmental Protection
Agency

Environmental Criteria and
Assessment Office
Research Triangle Park NC 27711

EPA-600/9-79-006
June 1979

Research and Development

EPA

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REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA 22161

EPA-600/9-79-006

June 1979

Suspended Particulate Matter

A Report to Congress

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PREFACE

This report was written in accordance with Section 403(a)(1) of the Clean Air Act Amendments of 1977, P.L. 95-95, as noted under 42 U.S.C. Section 7548. The purpose of this report is to assess health and welfare effects caused by suspended particulate matter in relationship to size, weight, and chemical composition.

The report does not constitute an in-depth review of the original published literature; rather, it summarizes, through the use of recent reviews by well-known experts in the field, the current scientific position with regard to knowledge of airborne particles, their effects on public health and welfare, and the respective levels at which these effects are thought to occur.

The Agency is pleased to acknowledge the efforts of all persons and groups who have participated in preparing this document.

ABSTRACT

This report is in response to Section 403(a)(1) of the Clean Air Act Amendments of 1977, P.L. 95-95, as noted under 42 U.S.C. Section 7548. The report covers (1) a review of the physical and chemical characteristics of airborne particles (source, composition, and sampling site as related to size); (2) a review of the effects of particulate matter on public welfare (ecological, materials, atmospheric, aesthetic); (3) the status of human exposure to airborne particles as related to source; and (4) a review of the effects of airborne particles on human health (lung deposition, chemical composition, interactions, and potentiating conditions).

Although there is a wide divergence of opinion among experts and scientific groups with respect to the issues of particulates the following can be concluded from the available information:

1. A major portion of the adverse health and welfare effects of air pollution is due to airborne particles.
2. Although pollution levels have declined in many U.S. localities in recent decades, there is still need for improvement in several of our cities.
3. Additional research is needed to improve the scientific basis for future airborne particle standards as outlined by EPA (cf. Inhalable Particulate Research Plan, Project Officer, Dr. Roger Cortesi, EPA, ORD).

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1. EXECUTIVE SUMMARY

This document is submitted as a Report to Congress, mandated by Section 403(a)(1) of the Clean Air Act Amendments of 1977, P.L. 95-95, as noted under 42 U.S.C. Section 7548, concerning the health and welfare effects of airborne particle pollution. The report provides an extended summary of these effects primarily as described in the review by the National Academy of Sciences (NAS) (Airborne Particles),[50] but also as described in other recent reviews. These reviews are: (1) Health Effects of Particulate Pollution: Reappraising the Evidence. A. E. Bennett et al., 1978; (2) Environmental Health Criteria for Sulfur Oxides and Susperded Particulates. World Health Organization, 1978; (3) Fine Particulate Pollution. World Health Organization, 1977; (4) Intra-Urban Mortality and Air Quality: An Economic Analysis of Pollution Induced Mortality. J. Gregor, 1977; (5) Total Suspended Particulates: Review and Analysis. R. Wells, 1976; (6) Air Quality Criteria for Particulate Matter, National Air Pollution Control Administration, 1969. The report represents an interim summary of health and welfare effects associated with airborne particles. A comprehensive review of health and welfare effects and other aspects is currently being conducted as part of the revision of the particulate matter criteria document. An external review copy of the criteria document will be available by late 1979, with final publication by late 1980. The material in this interim report is summarized under several headings as indicated below.

Characteristics of Airborne Particles

Airborne particles include a variety of chemical substances distributed over a wide range of particle sizes. The distinction between "fine particles" [<2 micrometers, (μm)] and "coarse particles" (>2 μm) is a fundamental one.

There is now an overwhelming amount of evidence that not only are two modes usually observed in the mass or volume distribution of well-mixed urban and rural aerosols, but that the fine and coarse modes are normally quite different in chemical composition. The fine and coarse particle modes originate separately, are transformed separately, are removed from the atmosphere by different mechanisms, require different control techniques, have different chemical composition, have different optical properties, and exhibit differential penetration of the respiratory tract. Therefore, the distinction between fine and coarse particles is of fundamental importance to any discussion of the physics, chemistry, measurement, and respiratory tract deposition of aerosols.

The major components of the fine fraction of the atmospheric aerosol are sulfates, ammonium ions, hydrogen ions, condensed organic matter, and trace levels of lead. The fine fraction, as a percentage of total suspended particulate matter, varies from 20 to 60 percent in urban areas. The percentage of the fine particle fraction which is formed mainly from atmospheric photochemical reactions varies from 60 to 80 percent in these urban areas (percentages based on short-term, intensive studies). Also, several studies have shown that potentially toxic carcinogenic species, such as polynuclear aromatic compounds, arsenic, selenium, cadmium, and zinc, which can exist as vapors are more concentrated in the fine particle fraction.

Materials typically found in the coarse fraction include those produced from mechanical processes such as grinding operations; rubber tire particles and other road dust; and also wind-blown dusts, sea salt, and pollen.

Sources and Concentrations

Exposures to airborne particles are determined not only by the amount and type of emitted pollutants, local topography, and meteorological conditions,

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but also by one's residential or occupational proximity to sources of such pollution and the rate, depth, and mode (mouth or nose) of breathing. For those pollutants that can accumulate in tissues, such as lead, cadmium, and fat-soluble organic compounds, tissue levels can increase with duration and frequency of environmental exposures.

Although the mass emissions of airborne particles from natural sources are thought to exceed those from man-made sources, naturally derived particles are both quantitatively and qualitatively less significant to human exposures in the urban environment. As an example, it has been estimated that the natural particle component of the Los Angeles aerosol is less than 30 percent of the total. These natural particles (sea salts, dust, etc.) are also generally less hazardous to healthy individuals than are many of the particles from man-made sources. Indoor concentrations of airborne particles are sometimes as great or greater than outdoor concentrations, particularly in certain occupational settings and in homes in which cigarette smokers live. Typically, urban environmental concentrations greatly exceed those of rural locations. Urban particulate pollution increased by 18 percent from 1940 through 1970, principally due to stationary fuel combustion sources and industrial process losses. Subsequent to 1970, particulate levels began to decline. The most significant downward trends in emissions were 33 percent for stationary fuel combustion sources and 54 percent for industrial processes. These downward trends resulted from the change from coal to natural gas and oil, the adoption of local smoke control ordinances, and the adoption of national emission standards. This decreasing trend that has been observed from 1970 through 1977 does not imply that all components of total suspended particulate are decreasing. However, a number of toxicologically significant particulates

such as trace metals and polycyclic organic matter have decreased. Prominent among the metal components of total suspended particulate showing a decline are vanadium, manganese, and nickel. Nationwide lead averages have declined since 1972 due to the introduction of the lower compression engines and the use of lower lead gasoline. This trend should continue with the shift of usage from low-lead gasoline to no-lead gasoline.

Health Effects

There has been increasing emphasis on examining what particle size fraction is of significance for health impacts. Recently, the EPA evaluated available information on the disposition of particles in the human respiratory tract. The analysis is summarized in "Size Consideration For Establishing A Standard For Inhalable Particles." This analysis provides the theoretical basis for designation of a size-specific airborne particle standard that would provide for more effective control of those particles most likely to be responsible for adverse health effects. The analysis indicates that under the most sensitive conditions (mouth breathing) particles of up to 15 micrometers in diameter can penetrate to the upper portions of the lung (conducting airways). This fraction, termed inhalable particulate (IP) matter makes up roughly 50 to 60 percent of total suspended particulate matter and includes all of the fine (<2.5 μm) and some of the coarse (2.5 to 15 μm) mode fraction. Based on this analysis, a major expansion of air pollution monitoring and health studies was initiated in 1979 to monitor and evaluate the health effects of inhalable as well as fine particulate matter.

Past studies of particulate pollution have indicated that high levels of airborne particle pollution are correlated with increased incidence, prevalence and severity of respiratory diseases and with increased mortality from these

and other diseases. The minimum concentration and duration of airborne particle exposure at which these health effects become significant cannot be precisely specified at this time. Furthermore, due to the limitation of monitoring methods used in these historical studies, it is difficult to correlate health effects directly with particle size and chemical composition.

Various reviews of the available information, including a report by a task force of the World Health Organization (1979), have concluded either that levels approximating the present primary TSP standards are reasonable ones for protection of public health or that there is not sufficient justification to change these standards on the basis of present knowledge. Others have concluded that the standards are more stringent than can be justified on the basis of present knowledge. Additional research is needed on the chemical and physical properties of particles in order to define better the characteristics and magnitude of particulate exposures which are consistent with public health.

Welfare Effects

Airborne particles produce well-documented effects on public welfare which include adverse effects on building materials and other exposed surfaces, atmospheric conditions, and aesthetic conditions, as well as possible influences on global temperatures and climate. The aesthetic effects of particulate pollution include the soiling of fabrics, buildings, and other surfaces, cultural losses due to damage to exposed architectural and historic artifacts, and degradation of visibility. Airborne particles have been associated with increased cloud cover and precipitation and have been implicated in shifts in temperature and climate. Moreover, airborne particles can seriously damage vegetation, wildlife, and natural systems when removed from the atmosphere by snow and rainfall. Long-range transported fine sulfates

and nitrates acidify soils and surface waters; nitrates, organic, and metallic substances in fine particulates can accumulate to harmful levels in soil, water, and plants.

It can be concluded that (1) a major portion of the adverse health and welfare effects of air pollution is due to airborne particles; (2) although pollution levels have decreased in many localities in recent decades, there is still need for considerable improvement in some of our cities; and (3) additional research is needed to improve the scientific basis for future airborne particle standards.

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2. INTRODUCTION

Section 403(a) of the Clean Air Act Amendments of 1977, P.L. 95-95, as noted under 42 U.S.C. Section 7548, requires the Administrator of the Environmental Protection Agency, in cooperation with the National Academy of Sciences, to submit before February 1979, a report to the Congress "on (1) the relationship between the size, weight, and chemical composition of suspended particulate matter and the nature and degree of the endangerment to public health or welfare presented by such particulate matter (especially with respect to fine particulate matter) and (2) the availability of technology for controlling such particulate matter."

This report is submitted in fulfillment of the first part [403(a)(1)] of that requirement. The form of the report is as follows:

Following the Executive Summary and the Introduction, Sections 3 through 6 present brief statements of the current scientific position with regard to knowledge of airborne particles and their effects on public health and welfare. References to prior reviews and to research reports are provided for those readers who wish additional documentation or detail.

3. CHARACTERISTICS OF AIRBORNE PARTICLES

3.1 PHYSICAL CHARACTERISTICS OF AIRBORNE PARTICLES

3.1.1 Size of Ambient Aerosol Particles

3.1.1.1 Aerodynamic Equivalent Diameter --Inasmuch as airborne particles are quite variable in shape, density, and chemical properties, their size and weight are generally characterized by an indirect measure referred to as the aerodynamic equivalent diameter: the diameter of a spherical particle of density 1.0 which would fall through air in earth's gravitational field at the falling speed of the observed particle. Aerodynamic equivalent diameter is, therefore, a function of both shape and density and is not necessarily indicative of the actual diameter of the observed particle. References in this report to particle diameter refer to this measure.

3.1.1.2 Size Distribution--Suspended atmospheric particles range in diameter from a few nanometers (nm) to several hundred micrometers (μm).* The numerical distribution of particles within this range normally exhibits two or three maxima. The bimodal distribution, with maxima at about 0.3 and 10 μm and a minimum at about 2 μm, is most common; but trimodal distributions (with the third peak (nuclei mode) corresponding to particles with diameters less than 0.1 μm) are also observed in ambient aerosols. Two micrometers is often used as the approximate demarcation point between "fine" and "coarse" particles. Within the definition of fine particles two distinct populations can exist, the "Aitkin nuclei" (<0.1 μm), emitted as such or formed by vapor condensation,

---

* This corresponds to a range from approximately the ultramicroscopic diameter of the smaller viruses to the diameter of dust particles which are easily visible. A nanometer is one one-thousandth of a micrometer (micron).

and the "accumulation mode" particles (0.1 to 2 μm), formed chiefly by coagulation and condensation of smaller particles. In fresh emissions, the Aitkin nuclei are usually numerically predominant, whereas in well-aged aerosols most of the fine particles are in the accumulation mode. The ratio of mass and volume to particle size varies with the geographical location, the predominant type of pollution (fugitive dust or photochemical products) and the age of the aerosol. Examples of volume distribution curves for particles measured by several investigators at different locations are shown in Figure 1.[1] For more information on particle size, see Chapter 2 of the National Academy of Sciences document prepared for EPA titled Airborne Particles.[50]

3.1.1.2 Variation of Particle Size According to Source--To a large extent, the initial size of an airborne particle depends upon its source. Secondary particles, formed in the atmosphere by condensation and/or chemical reactions of gases (such as sulfuric acid or organic solvent droplets), are very small initially. Although they rapidly grow by coagulation, most of them remain in the submicrometer range, also referred to as the accumulation mode.

Combustion processes, including automotive operation, produce particles which are mostly less than 1 μm in diameter, including particles of soot, various organic materials, and lead salts from leaded gasoline combustion. Many other chemical processes, such as sulfuric acid manufacture, also produce submicrometer-sized particles. Other sources which produce substantial amounts of fine particle emissions include metallurgical processes, paper mills, cement plants, and asphalt plants.

Grinding processes and most natural processes usually produce predominantly large particles (>2 μm). However, there is no reason to assume that they do not also produce fine particles.

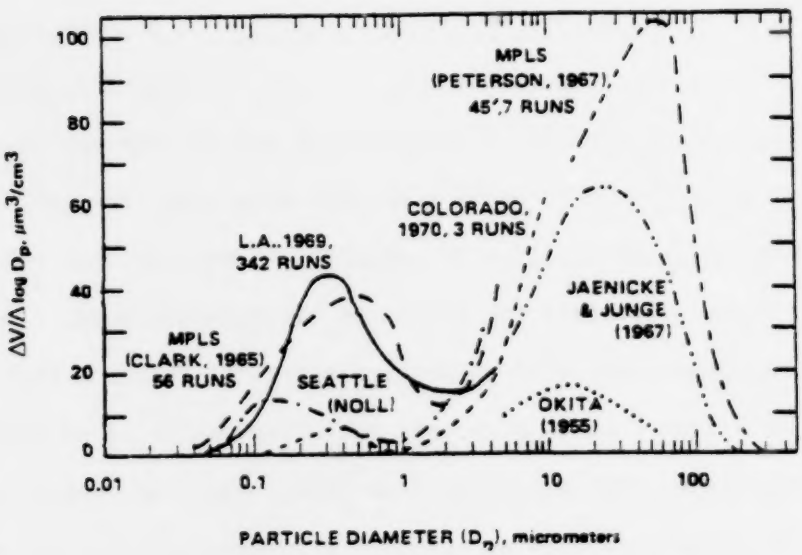

Figure 1. Comparison of volume distributions measured by several investigators in different locations.[1]

3.1.2 Physical State of Airborne Particulate Matter

Airborne particles, a phrase that is used interchangeably with "ambient aerosol," consist of both liquid and solid particles. Although these phrases are not normally understood to encompass particles of pure water, fine particles often serve as condensation or stabilizing nuclei and are associated mechanically with rain, fog, mist, and hail.

Aqueous droplets of dissolved or hydrated gases or solids are included conceptually within the definition of airborne particles. The fine particle fraction, especially, may contain a substantial liquid component at normal temperatures and humidities. Depending on the relative humidity, the liquid content of the fine particle fraction will vary considerably; for example, 50 percent of the fine particle fraction is water at 80 percent relative humidity.

Many of the fine particles (0.1 to 2 μm) are hygroscopic and, in some cases, deliquescent; and may change back and forth from the liquid to solid state with changes in ambient humidity. Solution/solid droplets of this nature include those of ammonium sulfate, sodium sulfate, sodium chloride, and ammonium nitrate.[50]

Other hygroscopic particles, while not undergoing a phase change, will exhibit changes in size with changes in humidity. Examples are particulate sulfuric acid, glycols, sugars, organic acids, and alcohols. Sulfuric acid and ammonium bisulfate are aqueous solution droplets at all relative humidities from 30 to 100 percent.[50]

Because the true mass of aqueous particles is not detected by methods which weigh or measure reflectance of filter-collected particle samples, water is often exempted from mass measurements of particulates. Therefore, as noted

above, measurements of airborne particles by these methods will underestimate both the mass and content of aerosols containing, for example, aqueous solutions or volatile or gaseous materials such as soluble organic compounds, nitric acid, or ammonia.[2] However, as noted above, the reverse error--the *in situ* formation of spurious particles on the collection filter due to chemical and physical interaction of the filter with atmospheric gases--also occurs and has been cited as a source of overestimation of the sulfate and nitrate content of ambient particles.[3,4,50] For example, two types of filters in common use were reported to collect 10 to 20 times as much artifactual as ambient particulate nitrate.[4]

3.1.3. Primary and Secondary Airborne Particles

Primary particles are those that are emitted as such; secondary particles are those formed in the atmosphere. Many of the ambient air particles are "secondary" aerosols: particles which are formed from materials (such as sulfur oxides and organic vapors) which are emitted as gases but which, upon cooling or upon chemical reaction within the atmosphere, condense to form particles. It has been estimated that secondary particles constitute as much as one-half of urban aerosols and 25 percent of the particulate pollution in the nation as a whole.[5]

Primary particles (those emitted as particles) are both coarse and fine, while most but not all secondary particles are in the "fine" size range.

3.2 CHEMICAL CHARACTERISTICS OF AIRBORNE PARTICLES

The specific chemical composition and characteristics of any atmospheric aerosol sample will depend upon the sources of the particles and on any reaction or selection that occurs during the residence time of the particle in the

atmosphere. Ambient aerosols are typically mixtures of greatly diluted emissions from numerous sources and are collected (or inhaled) after varying periods of settling, atmospheric mixing, atmospheric chemical reactions, and exposure to atmospheric humidity and washout. Therefore, the chemical characteristics of different samples can be extremely variable and complex.

Some of the chemical characteristics of ambient particles which are environmentally significant are composition, water solubility, acidity, hygroscopicity, deliquescence, efflorescence, and toxicity of both the atomic and molecular components of the particle.

3.2.1 Composition

Chemical categories of airborne particles which are of particular concern are toxic (including carcinogenic) organic compounds, inorganic fibers, toxic metals, and inorganic acids. To a large extent, chemical analyses of aerosols have determined atomic rather than molecular identities. A summary of the composition of ambient aerosol samples determined by the National Air Sampling Network is given in Table 1.[6-8] It can be seen that, of the components listed, sulfate is present in greatest amount, followed in order by nitrate anion. Of the metals, average concentrations were highest for iron and lead.

Acidity of airborne particles derives primarily from the presence of sulfuric acid, which is formed chiefly as secondary particles by oxidation of emitted sulfur oxides. Nitric acid, which is also present, is generally in the vapor state. Some of the sulfuric acid is neutralized by atmospheric ammonia and other cations. Sometimes, however, it remains partially unneutralized even in the presence of more than enough ammonia for theoretical neutralization.[9] It has been hypothesized that coating of the acid particles by organic material protects them against neutralization.

TABLE 1. GEOMETRIC MEAN[a] AND 90th PERCENTILE URBAN PARTICULATE CONCENTRATIONS IN THE UNITED STATES, 1971-1975

Pollutant	Concentration, $\mu g/m^3$	
	Geometric mean	90th percentile[b]
Suspended particulates[c]	~61	~97
Fractions:[d]		
Nitrates[e]	2.56	6.16
Sulfates	8.42	18.36
Ammonium (salts)	0.05	0.91
Beryllium	L.D.[f]	0.08
Cadmium	L.D.	0.007
Chromium	0.006	0.140
Cobalt	L.D.	LD
Copper	0.114	0.37
Iron	1.18	2.32
Lead	0.86	1.78
Manganese	0.03	0.92
Nickel	L.D.	0.034
Titanium	0.032	0.08
Vanadium	L.D.	0.07

[a]Based on 50th percentile

[b]90th percentile of quarterly composite measurements.

[c]Represent 1792 sampling sites.

[d]Represents 300 of the 1,792 Hi-Vol sampling sites.

[e]Possibly high by 70 to 90% due to artifacts.

[f]L.D. - Less than discrimination limit.

3.2.2 Composition as Related to Size

There are certain correlations between particle size and chemical composition, as indicated by the two summaries given in Table 2.[10-20] Sulfate, nitrate, elemental carbon, condensed organic vapors, ammonium ion, and lead occur chiefly in the fine-particle fraction and make up the major part of that fraction. In six U.S. cities, the mean diameter of particles containing metals ranged from a low of 0.4 to 0.7 μm for lead and vanadium to a high of 2.4 to 3.5 μm for iron. Particle diameters for polycyclic organic matter were found by another study to range from 0.3 to 0.5 μm.[50]

Various metals, as indicated by Table 2, plus alkaline particles and plant parts, occur chiefly in the coarse fraction. This fraction consists largely of mechanically produced particles such as soil, road and tire dust, and rock dust.

3.2.3 Effect of Sampling Method on Measured Particle Pollution --The two classical and historically most significant approaches to measuring the mass of airborne particles are based on gravimetric measurement of particles collected on a filter (Hi-Vol Sampler) and on optical transmission or reflectance of particles collected on filters (British Smoke Shade and AISI tape samplers). The British Smoke Shade method* measures the degree of blackening of a filter through which a low velocity (0.05 ft^3/min) stream of ambient air has been drawn for a specified length of time. The mass of particles accumulated on the filter is measured in terms of reflectance and converted to a mass basis by means of a "standard smoke" calibration and ultimately reported in micrograms per cubic meter.

The gravimetric method used most commonly in the United States has been the High-Volume (Hi-Vol) Method. Particles from a high-velocity (40 to 60

*Hereinafter referred to as the Smoke Shade method.

TABLE 2. CORRELATIONS OF CHEMICAL CONTENT WITH PARTICLE SIZE

a) Predominant Particle Size for Various Substances[10-19]

Normally fine	Normally coarse	Normally bimodal	Variable
Sulfates	Iron	Chloride	Nickel
	Calcium		
Organic (condensed vapors)	Titanium	Nitrate	Tin
	Magnesium		Vanadium
Lead	Potassium		Antimony
Arsenic	Phosphate		Manganese
Selenium	Silicon		Zinc
Hydrogen ion	Aluminum		Copper
Ammonium salts			
Soot			

b) Ratios of Element Distribution Between Fine and Coarse Particles[20]

(St. Louis Urban Aerosol, 18-day average, Aug.-Sept., 1975)

Predominantly fine		Predominantly coarse	
Element	Fine/coarse	Element	Fine/coarse
Sulfur	8.90	Calcium	0.09
Lead	3.67	Silicon	0.13
		Iron	0.29
		Potassium	0.33
		Titanium	0.55

ft^3/min) ambient air stream are collected on a filter and weighed. Larger particles, including blowing dust particles, can be entrained in the rapid air stream and collected by this method. Therefore, except at very high levels of fine particle pollution, and assuming constant sampler flow rate, measurements of ambient particulate matter typically give higher values by Hi-Vol than by the Smoke Shade method. The disparity is greatest at lower levels of particulate pollution (when coarse particles represent a larger fraction of the Hi-Vol sample). For example, an urban pollution Hi-Vol measurement of 200 $\mu g/m^3$ was reported to correspond to a Smoke Shade reading of 100 $\mu g/m^3$, while at 500 $\mu g/m^3$ the two methods yielded approximately equal results.[21] However, with unusually dark particles, the Smoke Shade method can give values higher than the corresponding gravimetric values.[22] Markedly varying particulate concentrations during the sampling period may result in Hi-Vol sample biases because the collection efficiency decreases non-uniformly over the sampling interval as more particles are deposited on the filter (Figure 2). Underestimation of particulate levels can result under conditions of constant TSP levels (Figure 2A) and under conditions of increasing TSP levels (Figure 2B). If particulate levels are declining (Figure 2C), an overestimation can result.

It should be kept in mind (1) that the Smoke Shade Method can under- or overestimate the collected particulate matter depending upon whether the particles are unusually optically light or dark since this method depends on reflectance, whereas gravimetric methods are color-independent; (2) that some overestimation of particle mass by the Hi-Vol method will occur due to interaction of the acid gases (i.e., SO_2, NO_2, HNO_3) with the alkaline surface of the glass fiber filter;[3,4] and (3) that both methods will fail to adequately quantify the mass of those particles that consist of air pollutants dissolved or suspended in or on aqueous droplets.

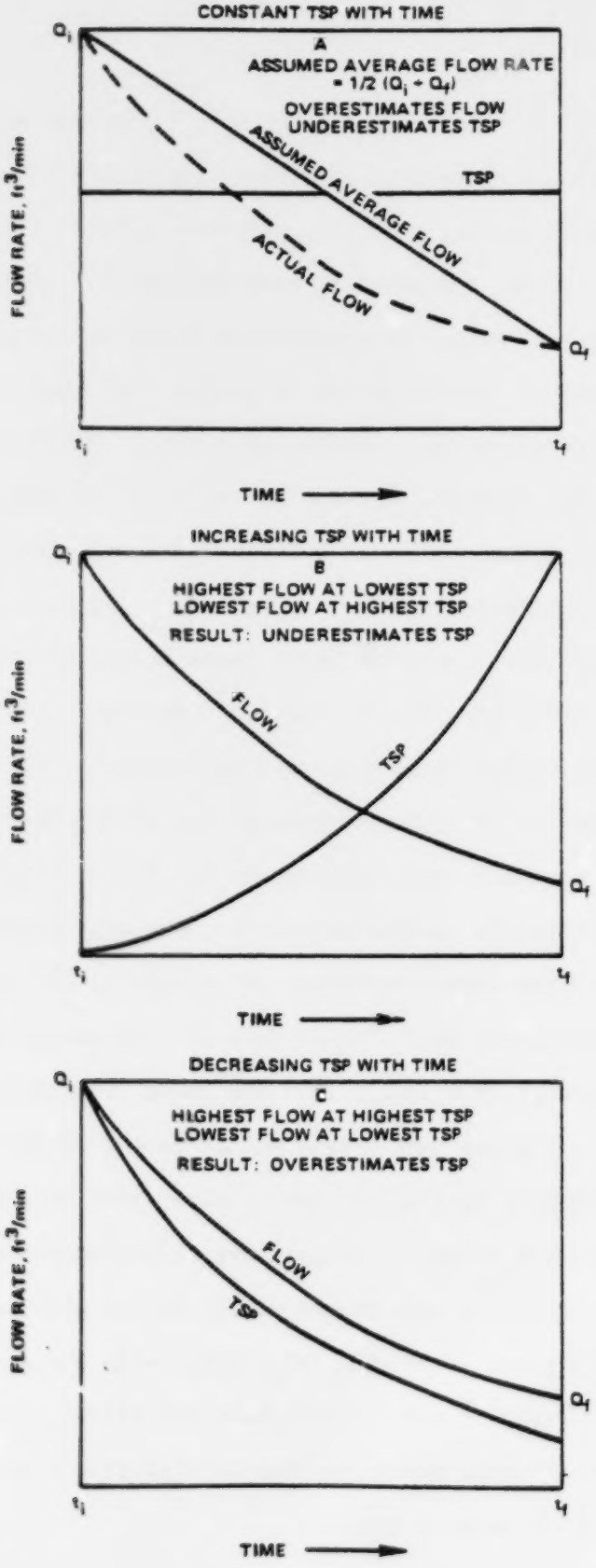

Figure 2. Estimation of TSP levels by the Hi-Vol method under varying conditions of particulate pollution.

In addition to the above instruments that have been used routinely in the United States and England for measuring the total mass of aerosols, impactors afford the additional option of separating the airborne particles according to size for subsequent chemical analysis. Impactors that separate aerosol particles depend upon the relative balance between inertial and aerodynamic forces. In the conventional type of aerosol impactor (e.g., cascade), an airstream turns abruptly as it approaches a flat surface. Particles with the largest inertia tend to maintain a straight trajectory and impact on the surface, whereas smaller particles will follow the air streamlines and not be collected. Inertial impactors such as these have been used extensively to collect airborne particles (e.g., CHESS data); however, difficulties with particle bounce, reentrainment, and non-uniform deposition have limited their use. To reduce particle bounce error the impaction surface may be coated with a non-volatile grease; however, large weighing errors are often encountered using greased surfaces. The problem inherent in collection by impaction onto surfaces is eliminated by using a "virtual" impactor in which particles are impacted into a slowly pumped void and in turn pulled onto a filter. The dichotomous "virtual" impactor as described by Stevens and Dzubay[85] was designed with an aerosol inlet such that the instrument fractionated particles into <3.5 μm and >3.5 μm with an upper limit of 20 μm. In order to provide a data base for evaluating health effects of fine and inhalable particles, the dichotomous sampler and the aerosol inlet were modified to give cut points of 2.5 μm and 15 μm, respectively. These particles are to be collected from air samples and deposited on Teflon filters with a 1.0 μm pore size. The monitoring network describing the collection of these inhalable particles has been described by Rodes in an Environmental Science and Technology article by Miller.[86]

4. AEROSOLS: SOURCES AND CONCENTRATIONS

4.1 SOURCES AND EMISSIONS

4.1.1 Global

Particulate air pollution sources can be categorized as man-made or natural. Man-made atmospheric particles are formed by condensation of emitted gasses, by chemical reactions (including combustion), or by mechanical pulverization. Natural sources include such processes as volcanic action, ocean spray, dusting of soil, forest fires, and the release of particulate organic matter from vegetation. Reentrainment of dust originating from both natural and man-made sources occurs when previously deposited particles are resuspended by air movement or other processes.

Global estimates of particulate emissions are presented in Table 3 for both man-made and natural sources.[23] This estimate, as well as those by Robinson and Robbins[24] and Hidy and Brock[25] show that on a global scale anthropogenic particulate sources constitute from perhaps 5 percent to 50 percent of the total, regardless of particle size and age.[50] Such estimates of global aerosol budgets include both primary and secondary particulate; however, they do not distinguish between fine and coarse particles and cannot be applied directly to urban regions. Most aerosol in urban regions is clearly dominated by emissions from human activity, particularly by the production of sulfur dioxide from the combustion of oil and coal. Miller et al.[26] developed an approach to an urban budget for the Los Angeles basin showing the relative importance of human and naturally produced particulate matter, which includes both primary and secondary particles.

TABLE 3. GLOBAL ESTIMATES OF PARTICLES SMALLER THAN 20 μm RADIUS EMITTED INTO OR FORMED IN THE ATMOSPHERE[23]

Type of particles		Number in atmosphere, 10^9 kg/yr
Natural		
Soil and rock debris[a]		100-500
Forest fires and slash-burning debris[a]		3-150
Sea salt		(300)
Volcanic debris		25-150
Particles formed from gaseous emissions		
Sulfate from H_2S		130-200
Ammonium salts from NH_3		80-270
Nitrate from NO_x		60-430
Hydrocarbons from plant exudations		75-200
	Subtotals	773-2,200
Manmade		
Particles (direct emissions)		10-90
Particles formed from gaseous emissions		
Sulfate from SO_2		130-200
Nitrate from NO_x		30-35
Hydrocarbons		15-90
	Subtotal	185-415
	TOTAL	958-2,616

[a]Includes unknown amounts of indirect man-made contributions.

4.1.2 Nationwide

Table 4 provides historical data on nationwide estimates of total particulate emissions in the United States from 1940 to 1970.[27] These data, derived from major source categories of primary particles, indicate an average increase of about 0.6 percent per year, from 20.7 to 24.6 million tons. This represents an overall increase of approximately 18 percent attributed to anthropogenic sources, primarily fuel combustion in stationary sources and industrial process losses. Table 5 indicates that since 1970 estimated emissions have decreased due to increased use of particulate control devices, even though fuel use and industrial output have increased substantially. Recent emission estimates, based on 1977 emission factors, show that total emissions from transportation have essentially remained constant throughout the 1970-1977 period.[28] This constant trend is the net effect of the decrease in the amount of particulate discharged per mile traveled (emission rates) and the increase in vehicle miles traveled. In other source categories, namely stationary fuel combustion and industrial processes, there were significant downward trends of 33 percent and 54 percent, respectively.

Estimates for natural sources of particulate far outweigh those attributed to man on a global scale; however, on a nationwide basis estimates do not include those particles that are formed in the atmosphere by photochemical reactions, namely, secondary particles (e.g., sulfates). On a nationwide basis the effective manmade impact of secondary particles is extremely difficult to quantify.

4.1.3 Variation of Chemical Composition with Source

Although it is not generally possible to identify in detail the specific sources of an ambient aerosol, there are characteristic chemical differences

TABLE 4. NATIONWIDE ESTIMATES OF PARTICULATE EMISSIONS, 1940-1970[a]
(10^6 tons/year)

Source category	1940	1950	1960	1970
Fuel combustion in stationary sources	9.6	9.0	7.6	6.8
Transportation	0.4	0.4	0.5	0.7
Solid waste disposal	0.4	0.6	1.0	1.4
Industrial process losses	8.8	10.8	11.9	13.3
Agricultural burning	1.6	1.8	2.1	2.4
Miscellaneous	6.4	3.3	2.1	1.0
Total	27.1	25.9	25.3	25.6
Total controllable[b]	20.7	22.6	23.2	24.6

[a]Based on 1970 emission factors.

[b]Miscellaneous sources not included.

TABLE 5. RECENT NATIONWIDE EMISSION ESTIMATES 1970-1977

(10^6 metric tons/yr, expressed as TSP)

Source Category	1970	1971	1972	1973	1974	1975	1976	1977
TRANSPORTATION	1.2	1.1	1.2	1.2	1.2	1.1	1.1	1.1
Highway vehicles	0.7	0.7	0.8	0.8	0.8	0.8	0.8	0.8
Nonhighway vehicles	0.5	0.4	0.4	0.4	0.4	0.3	0.3	0.3
STATIONARY FUEL COMBUSTION	7.1	6.6	6.4	6.5	5.6	5.0	4.6	4.8
Electric utilities	4.1	4.0	4.1	4.4	3.8	3.7	3.3	3.4
Industrial	2.6	2.2	2.0	1.8	1.5	1.1	1.1	1.2
Residential, commercial and institutional	0.4	0.4	0.3	0.3	0.3	0.2	0.2	0.2
INDUSTRIAL PROCESSES	11.9	11.3	10.6	10.3	8.9	6.5	6.2	5.4
Chemicals	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Petroleum refining	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Metals	2.1	1.9	1.9	2.1	1.9	1.4	1.5	1.3
Mineral products	7.8	7.4	6.9	6.4	5.5	3.7	3.2	2.7
Oil and gas production and marketing	0	0	0	0	0	0	0	0
Industrial organic solvent use	0	0	0	0	0	0	0	0
Other processes	1.6	1.7	1.5	1.5	1.2	1.1	1.2	1.1
SOLID WASTE DISPOSAL	1.1	0.8	0.7	0.6	0.6	0.5	0.5	0.4
MISCELLANEOUS	0.9	1.1	0.7	0.6	0.7	0.6	0.8	0.7
Forest wildfires and managed burning	0.5	0.7	0.5	0.4	0.5	0.4	0.6	0.5
Agricultural burning	0.3	0.2	0.1	0.1	0.1	0.1	0.1	0.1
Coal refuse burning	0.1	0.1	0	0	0	0	0	0
Structural fires	0	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Miscellaneous organic solvent use	0	0	0	0	0	0	0	0
TOTALS:	22.2	20.9	19.6	19.2	17.0	13.7	13.2	12.4

NOTE: A zero in this table indicates emissions of less than 50,000 metric tons/yr.

in emissions from different sources. Some of these chemical entities are summarized in Table 6, which includes the results of rough emission estimates from a number of reports.

Mobile sources are the major emitters of airborne lead (88 percent of total lead emissions), most of it as chlorides and bromides.[29] Vehicles are also responsible for 38 percent of total anthropogenic hydrocarbon emissions,[30] some of which are particulate or form secondary particles. Mobile sources release less than 2 percent of the sulfur oxides emitted.[31]

Stationary fuel combustion sources emit particles containing a number of substances of concern, including polycyclic organic matter (POM) and other organic material, carbon (soot, which is mainly of health significance because of its surface catalytic and adsorptive properties in relation to other pollutants), and certain metals. These sources also emit 81 percent of the national sulfur oxide emissions,[31] most of which are assumed to be converted to particles. Eighty percent of benzo(a)pyrene* emissions were estimated to come from coal combustion, as compared to only 4 percent from stationary oil burning and 2 percent from mobile sources.[32] Coal combustion emissions are also distinguished by the fact that they include 88 percent of the nationwide beryllium emissions.[33] Stationary oil combustion, on the other hand, generates 83 percent of nickel and 90 percent of vanadium emissions. Therefore, it would appear that except in the vicinity of atypical local sources, four metals can serve as virtual identifying markers for combustion-linked airborne particles: lead for mobile sources, beryllium for coal, and nickel and vanadium for oil. Such source-characteristic differences have been used in the chemical balance method to

*Benzo-a-pyrene (BaP) emissions are virtually all in the particulate state after cooling to atmospheric temperature.

TABLE 6. EMISSION ESTIMATES FOR MAJOR SOURCES OF CERTAIN AIRBORNE PARTICLES

Pollutant	Mobile sources	Smelting, metal-lurgical	Stationary combustion, Coal	Oil	Other	Total emissions Year	Tons
	% of total emissions of pollutant						
Particles[a]	4	36	32	2		1972	19.8×10^6
SO_x[a,b]	2	13	60	13		1972	33.2×10^6
NO_x[a]	35	<1	19	7	22[c]	1972	24.6×10^6
Hydrocarbons[a,b]	59	<1	<1	<1	27[d]	1972	27.8×10^6
Benzo(a)pyrene[e,f]	2		80	4		1975	978
Metals[g]							
Beryllium		3	88	6		1970	170
Cadmium		43			52[h]	1970	2,200
Chromium		68	9		10[j]	1970	17,000
Copper		84	7			1970	14,000
Manganese		74	11			1970	18,000
Nickel		11	83			1970	7,300
Lead	88-93	2				1970	230,000
Titanium			83		10[i]	1970	52,800
Vanadium		1	9	90		1970	20,000

[a] Ref. 34, Tables 4.1 and 4.2.
[b] Emitted chiefly as gases but with substantial conversion to secondary particles.
[c] Other stationary fuel combustion.
[d] Evaporated solvents.
[e] Values stated by authors to be only approximate.
[f] Ref. 41.
[g] Ref. 33, Table 1.
[h] Incineration of radiators and plated metal.
[i] Pigment production and use.
[j] Refractory production.

prepare an emission inventory for particles collected in the Los Angeles area.[5,35]

Noncombustion stationary source emissions have identifiable components that are characteristic of the processes involved. Thus, sulfuric acid plants emit sulfuric acid mist; emissions from an open hearth furnace have been found to be 89 percent iron oxide; particles from cement plants are chiefly calcium oxide and calcium carbonate.[7] Smelters and other metallurgical processes emit metal-containing particles, including 84 percent of the nation's total copper emissions;[33] particles with a high proportion of bio-organic materials are emitted by such sources as flour mills, grain elevators, and paper/pulp mills; and aerosols collected near the sea will include substantial fractions of sea salts.

4.2 TRANSFORMATION AND TRANSPORT

The makeup of a mixture of particles changes with time after emission. Insofar as elapsed time is correlated with distance from the source, these changes will be reflected in the changes with distance. The most significant transformations with time and distance are: (1) Condensation of vapors to form secondary particles (which produces high concentrations, near the point of emission, of particles less than 0.1 μm diameter); (2) particle coagulation and growth (particle interactions occurring very rapidly to produce larger particles from the condensation particles in the "accumulation mode" (0.1 to 2 μm) which persist in the atmosphere for days and for great distances); (3) chemical transformations such as oxidation of sulfur oxides and photochemical smog formation (occurring relatively slowly, so that products of these reactions are higher at moderate distances than at the site of emission); and (4) fallout,

rainout, and other removal mechanisms (which cause a steady decrease in particle mass and in mean particle diameter with increasing time and distance).

Long-range transport of sulfate and other inorganic compounds[36] as well as of polycyclic aromatic hydrocarbons[37] has been reported. Even coarse particles can be transported over long distances if they are injected above the atmospheric mixing layer as by volcanoes or by air mass movements over dry deserts. These processes of aerosol transformation and transport are described in detail in Airborne Particles.[50] Their net effect is that at increasing distances from the source, the aerosol concentration decreases and is increasingly dominated by accumulation mode particles with a mean diameter in the range of 0.4 to 1 μm, the nuclei having been removed by coagulation and the coarse particles by sedimentation.

4.3 CONCENTRATIONS

4.3.1 Trends in Airborne Particle Pollution

The routine episodes of high levels of visible smoke pollution which characterized wintertime in some U.S. cities before World War II no longer occur. The extensive replacement of coal as a home-heating fuel and the enforcement of local smoke emission ordinances brought about gradual decreases from the earlier high levels of urban smoke pollution, whether from coal or other fuels.[50] Since the passage of the Clean Air Act of 1970 and promulgation of the ambient air quality standards for TSP (primary, 75 $\mu g/m^3$) and several other pollutants, particulate emissions are being further reduced. Seventy-two percent of the sites monitored showed lower airborne particle concentrations in 1976 than in 1970. Improvement has been more marked in the Northeast and Great Lakes regions than in the western areas of the country

where windblown dust is often a significant aerosol constituent. The general pattern of change between 1970 and 1976 was little change at sites where 1970 concentrations were low and improvements at sites with higher 1970 concentrations.

At 53 percent of the sites, concentrations in 1976 were greater than those of 1975. The areas showing this reversal of the previous downward trend were generally in the Southeast, Midwest, and West. These same areas had dry soil conditions in 1976 due to the drought experienced by large areas of the country at that time. It has been suggested that the resultant increase in wind-blown dust was the reason for most of the increase in 1976 TSP levels.[38] Nationwide trends in total suspended particulate pollution are summarized for 1970-1977 in Table 5.

The gradual decrease shown by TSP data gathered at 17 of the National Air Surveillance Network (NASN) stations during the period 1966 to 1977 is shown in Figure 3.

The TSP curves of Figure 3 are not representative of every component of airborne particulate matter. A recent analysis of BaP trends during the 10-year period of 1966 to 1975 was based upon 34 urban (24 coke ovens) and 3 rural sites. These trends for the urban sites (Figure 4) are consistent with previous results indicating a steady decline of BaP.

Table 7 shows the trends of atmospheric trace metal levels in U.S. urban areas from 1965 to 1974.[33]

Prominent among the metals showing a decline are vanadium, manganese, and nickel. Nationwide lead averages have declined since 1972 because of the lower lead content of gasolines sold in recent years.

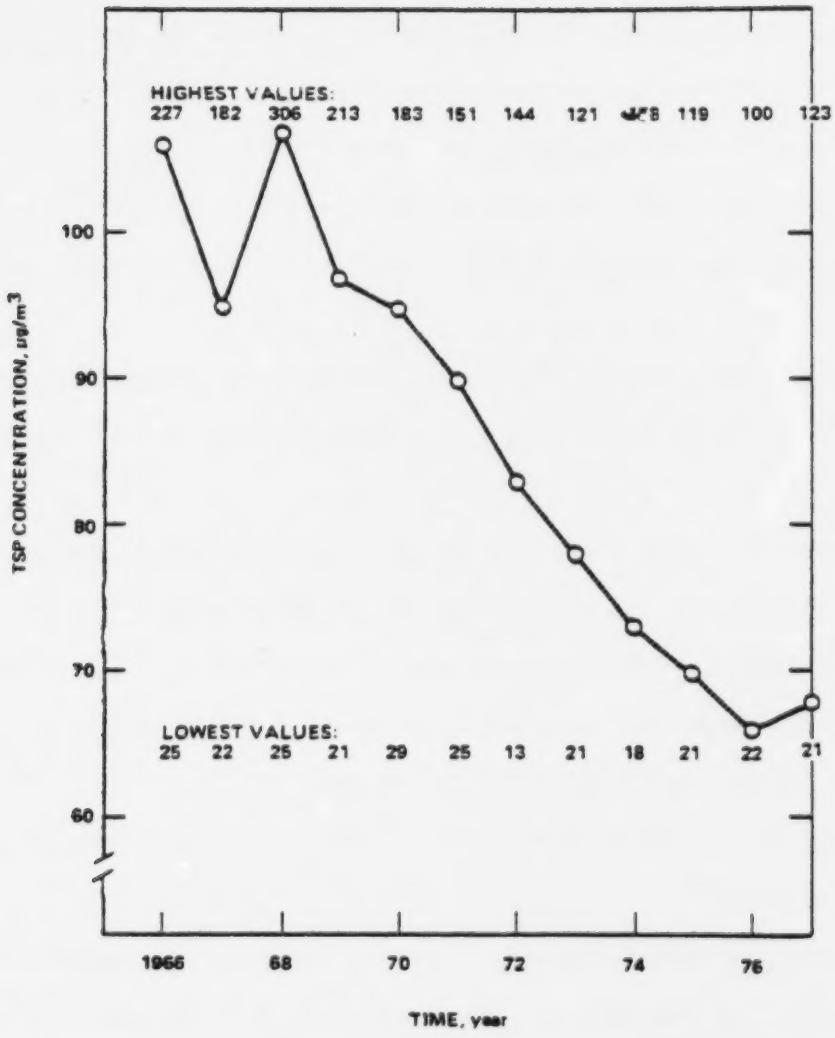

Figure 3. TSP annual means at 17 urban NASN stations.

NOTE: TSP means (EPA data) from all of those stations for which data were available for each of the years 1966-1977 were used to derive the composite averages shown. Stations included were Grand Canyon Nat. Park, Honolulu, Chicago, Indianapolis, New Orleans, Arcadia Nat. Park, Baltimore, Detroit, Cleveland, Toledo, Youngstown, Pittsburgh, Houston, Norfolk, Shenandoah Nat. Park, Seattle, and Charleston.

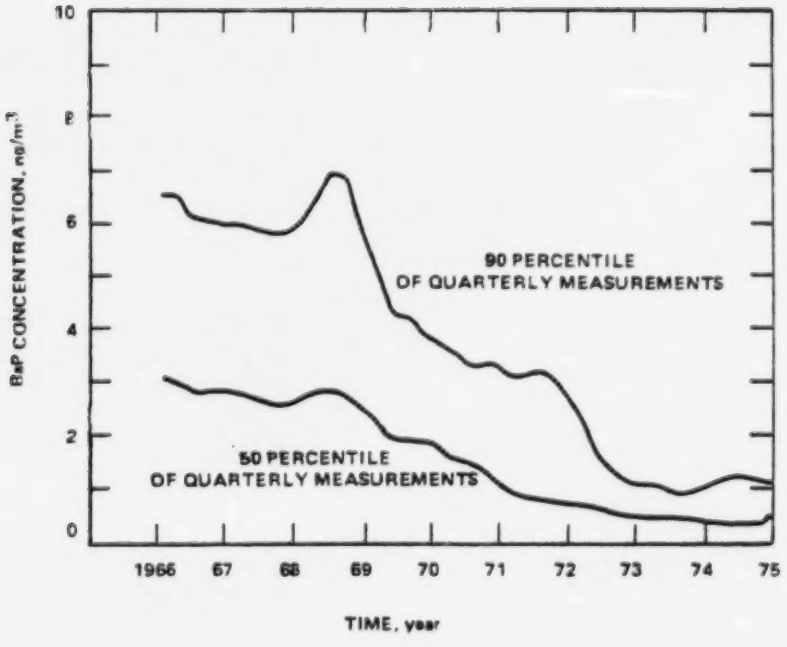

Figure 4. Benzo(a)pyrene seasonality and trends (1966 to 1975) in the 50th and 90th percentiles for 34 NASN urban sites.[63]

TABLE 7. TRENDS IN URBAN METAL CONCENTRATIONS AND THEIR POSSIBLE CAUSES[33]

Metal	Observed trends	Possible causes
Fuel-Combustion-Related Metals		
Beryllium	Unknown	--
Lead	Down last 5 years	Lower lead content in gasolines after 1969
Nickel	Down	Reduction of Ni in residual oils
Titanium	Up	Increasing use of coal in electric utilities
Vanadium	Down	Reduction of V in residual oils
Industry-Related Metals		
Cadmium	Down	Controls in metal industry and improved incineration practices
Chromium	No trend	Unknown
Cobalt	Unknown	--
Copper	No trend	Contamination from hi-vol commutator
Iron	Down	Improved incineration or waste burning practices, fuel switching, controls in steel industry
Manganese	Down	Controls in metals industry

4.3.2 Comparative Exposure to Airborne Particles

4.3.2.1 Urban Versus Rural Exposures

The generally higher exposures of urban residents to airborne particles is one illustration of the impact of source proximity. For example, data from the National Air Sampling Network in 1970 show that the mean annual average benzo(a)pyrene concentration was 1.9 ng/m^3 (range 0.1 to 19.3) at 120 urban stations versus 0.2 (range 0.1 to 0.4) for 20 nonurban stations.[31]

Such urban/rural differentials also exist for other constituents of airborne particles. The urban/suburban ratios for 22 trace elements in air of the Cleveland area are shown in Table 8. The median ratio is 2.2 and in no case did the suburban exceed the urban concentration.

Other comparisons of urban and nonurban aerosol exposures and the apparent effects of their differences on disease and death rates are presented and discussed in Airborne Particles.[50]

4.3.2.2 Indoor Versus Outdoor Residential Exposures

Because there is continual air exchange between interiors and exteriors of homes, outdoor aerosols--particularly the fine particles--will be included among the indoor airborne particles. However, this exchange also serves to dissipate airborne particles generated by indoor sources. Indoor sources include combustion processes such as those of cigarettes, furnaces, fireplaces, and stoves, and aerosol dispensers of consumer products. Although indoor concentrations of airborne particles, in the absence of smokers, are believed to be usually less than those outdoors, some studies have found higher indoor levels. It is recognized that the current emphasis upon improved insulation and weatherstripping for homes as an energy conservation measure could have either beneficial or adverse effects on indoor air quality, depending on

TABLE 8. RATIOS OF URBAN (U) TO SUBURBAN (S) CONCENTRATIONS IN AIR, CLEVELAND, OHIO AREA[36]

Element	U/S	Element	U/S	Element	U/S
Antimony	6.9	Mercury	3.0	Silicon, tin	1.8
Chloride	6.5	Iron	2.8	Copper, vanadium	1.8
Beryllium	6.1	Cadmium	2.5	Aluminum	1.7
Chromium	5.6	Sodium	2.4	Zinc	1.6
Cobalt	3.4	Magnesium	2.4	Arsenic	1.4
Bismuth	3.3	Manganese	2.2	Selenium	1.3
		Calcium	2.0	Bromide	1.2
		$\bar{X}$ = 2.8			

whether, as a consequence, the indoor air is more or less polluted than the outdoor air.

Three documents[39-41] recently prepared for EPA specifically address the problem of indoor air pollution, including indoor airborne particles. Because it is now known that indoor airborne particle concentrations sometimes are higher than the limits allowed outdoors by the ambient air quality standard while simultaneous outdoor concentrations are not, and because most individuals are exposed for longer periods each day to indoor than to outdoor environments, there is increasing concern as to the health impact of air quality in non-occupational indoor environments. Accordingly, it has been recommended that further monitoring of indoor air quality should be carried out and that epidemiological studies of relationships between indoor air pollution and health effects be conducted.[40] Some health studies such as the Harvard Air Pollution Health Study,[52] are already incorporating indoor monitoring.

4.3.2.3 Environmental Versus Occupational Exposures

In some occupational environments, the airborne particle concentration greatly exceeds that of the external environment. Examples are workplaces such as textile and paper mills, steel works, smelters, metallurgical processing plants, coke ovens, and mines.

These high occupational exposures frequently involve exposure to higher proportions of coarse particles than found in environmental exposures. They are of some importance to evaluating effects of particles because they may modify the response of an individual to airborne environmental pollution, and because they are the source of much of our information concerning the health effects of airborne particles.

5. EFFECTS OF AIRBORNE PARTICLES ON HUMAN HEALTH

5.1 DEPOSITION OF AIRBORNE PARTICLES IN THE LUNG

Adverse health effects of airborne particulate matter follow deposition of the particles in the respiratory tract. In man, the respiratory tract can be divided into three main areas on which experimental and theoretical research has been conducted on the deposition of inhaled particles. These areas are the upper respiratory tract extending from the anterior nares to the larynx, the conducting airways of the lower respiratory tract, i.e., the tracheobronchial region, and the gas-exchange areas. Beginning with the posterior portions of the nose, the nasal turbinates, and the trachea, the bronchial airways are ciliated and lined by mucus arising from glands and/or secretory cells. The nonciliated portions of the lung, which are lined by surfactant, are comprised of the respiratory bronchioles and the alveolar regions and represent the gas-exchange areas. Factors influencing respiratory deposition of particulate matter are discussed at greater length in Airborne Particles by NAS[50] and in Report on Inhalable Particles by the Environmental Protection Agency.[42]

Recently, the Environmental Protection Agency health scientists proposed a research program to develop information for a size-specific airborne particulate standard. This program will focus on "inhaled particulate" (IP) matter defined as airborne particles $\leq$15 μm aerodynamic equivalent diameter. This recommendation was based upon an increased deposition situation, i.e., mouth breathing with exercise, and relates to that fraction of particulate matter which can deposit in the conducting airways and the gas-exchange areas

of the human respiratory system. The analysis also recommended that a second particle size cut-point of $\leq$2.5 μm diameter be incorporated in the air sampling devices, based upon considerations of the chemical composition and the size distribution of airborne particles and on the predominant penetration of particles $\leq$2.5 μm diameter into the gas-exchange region of the respiratory tract. This analysis pointed out that data collected in this size range could be used in conjuction with epidemiological health parameters to refine an inhaled particulate standard in the future.

5.2 HEALTH EFFECTS OF GENERAL AIRBORNE PARTICLE POLLUTION

The present primary ambient air quality standard of 75 $\mu g/m^3$ for particulate matter is required on an annual basis for the protection of public health. In the NAS document entitled Airborne Particles,[50] scientific information concerning the health effects of airborne particles is described. A more concise review by one of the contributors to that volume has also been published.[43] Tabular summaries of evidence presented by different workers for particular health effects are provided in the statement by the American Thoracic Society[44] and in Airborne Particles.[50]

Exposure to airborne particles as it actually occurs almost always is accompanied by simultaneous exposure to other air pollutants and to other environmental chemicals. Although there is a tendency to regard these other pollutants as merely obstructing the view of the scientist seeking to detect the true effect of one particular pollutant, it must be kept in mind that the other pollutants are probably modifying the "true" effect of the pollutant studied and that the observed responses to the mixed but real exposure conditions are the ones that are relevant to human health. As an example, where

airborne particle pollution is coincident with sulfur oxides pollution, it is desirable to evaluate the health effect of these pollutants jointly.

There are indications that particles contribute more heavily than sulfur dioxide does to the health effects of this combination. For example, health indices in London have improved during a period (1950-1970) in which particulate pollution declined greatly along with a smaller reduction in sulfur dioxide. In any case, there is good evidence that excessive airborne particle/SO_2 pollution does cause illness and death, as well as increased prevalence, incidence, and severity of respiratory disease and symptoms. Incidence and progression of bronchitis and emphysema have been especially clearly linked to such pollution. The effects are most evident in the oldest and youngest age groups and in those people who are sick. There is also evidence of delayed appearance and of long-term persistence of the adverse effects of airborne particle exposures.[50]

The fact that urban residence is associated with increased incidence of cancer[45] and that mutagenic (Ames-test positive) components are extractable from urban airborne particles[46-49] suggest the possibility that airborne particles are also to some degree carcinogenic.

Table 9 provides estimated concentrations of particulates and sulfur dioxide that may affect health.[50] Estimates of adverse health effects attributable to sulfur dioxide, particulate sulfates, and total suspended particulates (TSP) are presented for short-term and long-term exposures, respectively, in Tables 10 and 11, reproduced from Airborne Particles.[50] Additional discussion of the evidence concerning this dose-response relationship is contained in Airborne Particles.[50]

TABLE 9. HEALTH EFFECTS AND DOSE/RESPONSE RELATIONSHIPS FOR PARTICULATES AND SULFUR DIOXIDE

Averaging time for pollution measurements	Place	Particles, mg/m^3	SO_2, mg/m^3	Effect	Reference
24 hour	London	2.00	1.04	Mortality	88
		0.75	0.71	Mortality	97
		0.50	0.50	Exacerbation of bronchitis	97
	New York City	6 COHS[a]	0.50	Mortality	94
		3 COHS	0.70	Morbidity	93
	Chicago	Not stated	0.70	Exacerbations of bronchitis	89
	New York City	0.145 (+?)	0.286	Increased prevalence of respiratory symptoms	95
	Birmingham	0.18-0.22	0.026	Increased prevalence of respiratory symptoms	95
	New York	2.5 COHS	0.52	Mortality	92
Weekly mean	London	0.20	0.40	Increased prevalence or incidence of respiratory illness	87
6 Winter months	Britain	0.20	0.20	Bronchitis sickness absence from work	101
Annual	Britain	0.07	0.09	Lower respiratory infection in in children	90
		0.10	0.10	Bronchitis prevalence	96
		0.10	0.12	Respiratory symptoms and lung function in children	99-100
	Buffalo	0.08	0.45[b]	Mortality	102-103
	Berlin	0.18	0.73[c]	Decreased lung function	91

[a] Coefficient of Haze Units.
[b] mg $SO_3/cm^2/30$ days.
[c] mg $SO_3/100\ cm^2/day$.

TABLE 10. THRESHOLD ESTIMATES FOR ADVERSE HEALTH EFFECTS ATTRIBUTABLE TO SULFUR DIOXIDE, PARTICULATE SULFATE, AND TOTAL SUSPENDED PARTICULATES (TSP), SHORT-TERM EXPOSURES

Adverse effect human health	Research approach	Type of estimate	Exposure level, $\mu g/m^3$			Duration	Safety margin, % contained in primary standard[a]		
			SO_2	Particulate sulfate	TSP		SO_2 std. 365 $\mu g/m^3$	Suspended sulfates (no std.)	TSP std. 260 $\mu g/m^3$
Mortality	Epidemiology	Worst case	30	No data	250	24 hr	0	No data	0
		Least case	500	No data	500	24 hr	37	No data	92
		Best judgment	300-400	No data	250-300	24 hr	0-9	No data	0-15
Aggravation of chronic lung disease	Epidemiology	Worst case	119	6	100	24 hr	0	No data	0
		Least case	500	No effect	>250	24 hr	37	No data	Less than 100
		Best judgment	365	10	>250	24 hr	0-37	0	0
Aggravation of asthma	Epidemiology	Worst case	23	6	75	24 hr	0	0	0
		Least case	>365	10	>260	24 hr	Less than 100	0	Less than 100
		Best judgment	180-250	8-10	100	24 hr	0	0	0
Aggravation of combined heart and lung disease	Epidemiology	Worst case	180	6	61	24hr	0	0	0
		Least case	>365	10-17	260	24hr	Less than 100	0	Less than 100
		Best judgment	365-500	8-10	70-100	24 hr	0-37	0	0
Irritation of respiratory tract	Epidemiology	Worst case	340	No data	170	2-3 days	0	No data	0
		Least case	340	No data	192	2-3 days	0	No data	0
		Point est	340	No data	170	2-3 days	0	No data	0

[a]Safety Margin = Effects threshold minus standard divided by standard x 100.

TABLE 11. THRESHOLD ESTIMATES FOR ADVERSE HEALTH EFFECTS ATTRIBUTABLE TO SULFUR DIOXIDE PARTICULATE SULFATES, AND TOTAL SUSPENDED PARTICULATES (TSP), LONG-TERM EXPOSURES

Adverse effect human health	Research approach	Type of estimate	Exposure level, $\mu g/m^3$			Duration, years	Safety margin, % contained in primary standard[a]		
			SO_2	Suspended sulfate	TSP		SO_2 std. 80 $\mu g/m^3$	Suspended sulfates (no std.)	TSP std. 75 $\mu g/m^3$
Excess mortality	Epidemiology	Worst case	120-198	No data	135	Unknown	50-148	0	80
		Least case	250	No data	175	Unknown	212	0	133
		Best judgment	250	No data	175	Unknown	212	0	133
Increase in prevalence of chronic bronchitis	Epidemiology	Worst case	50-90	96	60-100	3	0-12	0	0-33
		Least case	404	20	180	12	405	0	140
		Best judgment	95	14	100	5	19	0	33
Increased frequency or severity of acute respiratory illness in otherwise healthy families	Epidemiology	Worst case	91	9	75	1	14	0	0
		Least case	200	23	200	3	250	0	167
		Best judgment	91	9	100	3	14	0	33
Increase in family illness during influenza epidemics	Epidemiology	Worst case	106	14	126	1	33	0	68
		Least case	250	18	151	Unknown	212	0	101
		Best judgment	106	14	151	3	33	0	101

TABLE 11 (continued).

Adverse effect human health	Research approach	Type of estimate	Exposure level, $\mu g/m^3$			Duration, years	Safety margin, % contained in primary standard[a]		
			SO_2	Suspended sulfate	TSP		SO_2 std. 80 $\mu g/m^3$	Suspended sulfates (no std.)	TSP std. 75 $\mu g/m^3$
Increased lower respiratory tract infections in asthmatics	Epidemiology	Worst case	32	8	100	Unknown	0	0	33
		Least case	186	20	No	Unknown	133	0	---
		Best judgment	91	8	100	Unknown	14	0	33
Subtle decrease in ventilatory function	Epidemiology	Worst case	118-131	9	75-141	1	48-64	0	0-88
		Least case	400-500	28	200	9	400-525	0	167
		Best judgment	200	11	100	8-9	150	0	33

[a]Safety Margin = Effects threshold minus standard divided by standard x 100.

The severity of the health effects at any given airborne particle concentration and the lowest pollution level required to evoke significant adverse health effects in the most sensitive populations are not easily determined. With so many factors determining exposures (Table 12) and with so many variables influencing the response to a given exposure, any epidemiological study of dose/response relationships must necessarily cope with uncertainties in both the dose and response functions. Therefore, there is some disagreement among scientists as to the level at which airborne particle pollution becomes hazardous.

The conclusions of many of the studies cited in Tables 9-11 have been questioned by Bennett et al.[51] (report prepared for the American Iron and Steel Institute) generally on the basis of the fact that one or more relevant variables were uncontrolled. The extent to which the correlations of health effects with pollution indicated by such studies should be discredited must remain a matter of judgment. As is apparent from the above mentioned tabulation of related variables, it can be expected that there will be some uncontrolled variables in any epidemiologic study of the health effects of air pollution.

In addition to differing judgments as to the reliance that can be placed on various studies with their varying levels of imperfection, there are also sometimes differences of opinion as to how even such information as is available in a given study should be interpreted.

In addition to variations in the design and interpretation of various studies, other factors also undermine precise assessment of the epidemiological evidence. These include differences in methodology of airborne particle measurement; the possibility that health effects of exposure to air pollution may not appear or may persist until many years later;[53] the high mobility of the American population that causes the enviroment of a given population to be only partially representative of exposure of that population; the changing

TABLE 12. VARIABLES RELATED TO THE HEALTH EFFECTS OF AIRBORNE PARTICLES

Exposure factors	Response factors	Incidental factors	(Linkage factors[a])
1. Emissions (kind and amount)	1. Temperature	1. Income	(Exp. No. 6 - 10)
2. Synergistic combinations	2. Physical condition	2. Education	(Exp. No. 6 - 10)
3. Topography	3. Concurrent disease	3. Social class	(Exp. No. 6 - 10)
4. Climate, weather, humidity	4. Nutritional status?	4. Race[b]	(Exp. No. 6 - 10)
5. Seasonal and diurnal changes	5. Prior exposure	5. Sex[b]	(Exp. No. 7,9,10)
6. Place of residence	6. Smoking history	6. Sanitation	(Resp. No. 2,3)
7. Occupation	7. Age	7. Health insurance	(Resp. No. 2-4)
8. Duration of exposure		8. Access to medical care	(Resp. No. 2-4)
9. Smoking (active and passive)		9. Population density	(Resp. No. 3, Exp. No. 6)
10. Physical activity		10. Age[b]	(Resp. No. 2,5)
11. Breathing mode (mouth or nose)			

[a]Causal exposure or response factors, through which incidental factor may be linked to health effects of air pollution.

[b]Somewhat uncertain.

pattern and nature of air pollution; and changing social and behavioral patterns which modify the impact of such variables as race, sex, and smoking.

5.3 CONDITIONS WHICH MAY POTENTIATE HEALTH EFFECTS OF AIRBORNE PARTICLES

Associations between the health effects of airborne particles and correlated variables can be categorized as three types. A number of variables (which we will call exposure factors) can be assumed to influence causally the extent of exposure to airborne particle pollution; variables of a second type (response factors) directly influence the response of an exposed person to a given exposure; and variables of the third type (incidental factors), while correlated with pollution-related morbidity and mortality, are assumed to be non-causally linked to health effects through association with one or more of the exposure or response factors. Examples of some variables of each type are shown in Table 12.

The factors listed in Column 1 of Table 12 directly determine the exposure of a particular person to airborne particles. They include the nature and amount of pollutants emitted (No. 1-2), the geographic and atmospheric factors that determine the extent to which pollutants will accumulate in or be removed from the ambient air (No. 3-5), and the individual determinants that vary with personal behavior or circumstance (No. 6-11).

Some of these variables have been discussed in Sections 6.2 and 6.3.

A second category of factors (Column 2) will determine the response of each individual to a given level of airborne particle pollution. These response factors include external potentiating variables, such as temperature, and physiological factors.

Since low temperature alone can cause increased mortality in the elderly, low temperature may increase the vulnerability of the old or sick to superimposed

air pollution exposure. Studies have shown correlations of low temperature with airway resistance and with symptoms exhibited during air pollution episodes by bronchitis patients.[50] However, the extent to which the health effects of temperature and of air pollution are independent or interacting is not clear.

The physiological factors named in Column 2 (items 2 through 7) all relate to general physical condition and therefore to the capacity of the individual to resist pollution-imposed stress. There is abundant documentation to show that people with concurrent respiratory disease are especially vulnerable to air pollution and that, conversely, air pollution has decided impact on the incidence of respiratory disease. Many epidemiologic studies have taken advantage of these relationships by monitoring the symptomatic reaction of respiratory disease patients or the prevalence of respiratory disease during air pollution episodes. The quantitative aspects of these correlations, especially the minimal level of air pollution likely to evoke an adverse respiratory response, are still under debate.

The different responses of different age groups to air pollution are also well documented. Although the increased vulnerability of the elderly to air pollution may be in part related to increased cumulative exposures to prior air pollution, it is presumably chiefly due to the decreased physical fitness in older age groups with resultant increases in susceptibility both in disease and to air pollution. There is also evidence that children and especially infants are also more vulnerable to air pollution.[50]

The third column of Table 12 lists some incidental factors: variables which appear to be correlated with pollution-related morbidity or mortality

but which are categorized here as being thus linked only indirectly, through one or more of the more direct exposure or response factors. Possible causal linkages are indicated in Column 4. We have suggested that factors 1-5 can most reasonably be assumed to be linked through exposure factors while correlations of 6-10 with health effects seem more likely to be mediated by response factors.

The linkage of income level, for example, with pollution-related health effects can reasonably be seen as due to the decreased occupational and residential exposures to air pollution which increased income can command. Protective effects related to education, race, social class, and sex are also postulated to be mediated chiefly through variations in exposure. Such categorization of race and sex as indirect or non-causal factors is somewhat uncertain. However, there is no clear evidence of direct effects of these variables on response to air pollution unmediated by other exposure or response factors.

5.4 HEALTH EFFECTS OF SPECIFIC CHEMICAL COMPOUNDS

Airborne particles can be of health significance because some of their chemical components are potentially toxic per se and also because less hazardous particles, on interaction with other air pollutants or constituents, can be transformed into more hazardous ones.

5.4.1 Intrinsically Injurious Particles

A given inhaled particle may exert an adverse health effect either through chronic local tissue damage by an insoluble particle deposited in the deep lung or through solution and absorption of toxic components of the particle. Detailed understanding of the active agents and mechanisms which produce the adverse health effects is still largely non-existent. However, several chemical

components of the ambient aerosol are known or suspected to be contributors to these effects, either because of epidemiological observations or because of other information available from experimental, occupational, or clinical studies.

5.4.1.1 Fibers--Asbestos was first recognized as an occupational health hazard responsible for the condition known as pulmonary asbestosis and later as an occupational carcinogen when retrospective epidemiological studies showed that occupational exposure is strongly associated with malignancy of the lung, pleura, peritoneum, and gastrointestinal tract. The effects typically become manifest 20 or more years after exposure. There is increasing evidence that the general population may also suffer from inhalation exposure to asbestos, since asbestos fibers are found in the ambient air and pulmonary asbestos bodies are found in post-mortem examination of a large proportion of urban residents.[54] To protect the general population from exposure to high ambient levels of asbestos, the EPA promulgated an emission standard in 1973 which curtailed asbestos emissions from specific industries. The projected total reduction by 1977 from implementation of the standard was 93 percent.

5.4.1.2 Toxic Metals--Several metals which are known to be associated with airborne particles and which are known to be toxic are of concern.

Lead is the air pollutant metal which, on a nationwide basis, is believed to pose the greatest hazard. Airborne lead, at least 88 percent of which is from combustion of leaded gasoline, has been associated with increased lead concentrations in the blood of children and adults.[29] Primary exposure to airborne lead occurs from inhalation, while secondary exposure may occur through ingestion of foods and nonfood items which are contaminated by airborne lead. In people, lead affects the erythrocytes, the central and peripheral

nervous systems, soft tissues (kidney, liver), and bone. The latter sequesters 95 percent of the body burden of lead. Irreversible brain damage is one of the observed effects of overt lead poisoning in infants and children.

Biological effects which have been correlated with blood lead levels include elevated erythrocyte protoporphyrin and mild anemia (15 to 40 μg of lead/dl blood), peripheral nervous disorders and central nervous system damage (50 to 60 μg/dl), and severe neuro-behavioral impairment (80 to 120 μg/dl), sometimes resulting in convulsions and death. The minimum "safe" level of exposure to lead, as manifested by blood lead levels, is apparently variable among individuals. Formerly, blood lead concentrations less than 40 μg/dl were considered harmless. However, it has now been documented that hematological effects do occur at lead concentrations as low as 10 μg/dl.[29] Association of relatively low ($\leq$40 μg/dl) blood lead levels with learning deficits and with lower IQ scores has also been reported.[55] It is apparent that current airborne lead levels may be having undetected but quite serious effects on young urban residents. (NASN data for 1974 indicate that lead concentrations were less than 0.5 $\mu g/m^3$ at all nonurban stations but were 1.0 or higher at 32 percent of the urban stations.) Concern about the effects of airborne lead has prompted the EPA to propose and promulgate an air quality standard for lead based on health considerations. The present lead standard (1.5 $\mu g/m^3$ averaged over a calendar quarter) has as its goal a population mean blood lead level of 15 μg/dl or less among children who do not receive significant exposure to lead-based paints. Such an average would ensure blood lead levels not exceeding 30 μg/dl for 99 percent of such children.

Mercury is a highly toxic metal which occurs in air both as particles and vapor. Toxicity is due to accumulation in nerve tissue which ultimately can

lead to insomnia, loss of memory, and severe neurobehavioral and personality changes. Children and the fetus are the most susceptible to mercury poisoning. Release of mercury into the atmosphere from natural sources far outweighs the release from the major man-made source, coal combustion. On the average, the ambient concentrations in the large industrial cities are well below the 1 $\mu g/m^3$ level, and toxic airborne exposures are unlikely except in the vicinity of point sources.[56]

Cadmium accumulates in the body, especially in the kidney, where it has a very long biological half-life. Cadmium dust, when inhaled at high concentrations, can lead to severe kidney and lung damage. In view of the cumulative nature of this metal and of the resulting health effects, it seems appropriate to minimize the exposure of the general population. The average concentration in the ambient air is extremely low (approximately 0.002 $\mu g/m^3$) but may reach levels of 0.3 $\mu g/m^3$ near industrial point sources such as smelters.[57] The EPA is presently evaluating what type of regulatory action, if any, is needed to protect public health.

Arsenic is emitted mainly by metal smelters, coal combustion facilities, and the pesticide industry. In air, arsenic occurs mainly in particulate form. Approximately 80 percent of absorbed arsenic is retained and widely distributed in the tissues. Arsenic may cause both acute and chronic poisoning, but acute poisoning is rare. Both subacute and chronic poisoning can result from polluted air. Effects of arsenic poisoning include skin and mucous membrane abnormalities, gastrointestinal and nervous symptoms, and disorders of the circulatory system and the liver. In spite of the epidemiological association between exposure to arsenic and cancer of the skin, lungs, and

liver, a suitable animal model has not been developed that demonstrates carcinogenicity of arsenic. The epidemiologic studies involved exposure not only to arsenic compounds but to high levels of sulfur dioxide (at smelters) and of other heavy metals (at power plants). NASN reported annual average concentrations of arsenic in air ranging (in 1964) from nondetectable levels to 0.75 mg/m^3, with an overall average of 0.02 $\mu g/m^3$. The EPA is presently considering listing arsenic as a hazardous air pollutant.[58]

Beryllium is considered to be one of the most toxic metals in industrial use. The major emission source is coal combustion (88 percent of total emissions). All acute cases of beryllium disease have been associated with beryllium either in occupational settings or in communities adjacent to point sources. The Committee on Toxicology of the National Academy of Sciences concluded that a 30-day average concentration of 0.01 $\mu g/m^3$ in ambient air was a proven safe level since no cases of beryllium poisoning from exposures at or below that level were reported.[59] Beryllium is also reported to be carcinogenic,[60] although the point is presently being contested. Quarterly composite NASN data for 1970-1975 show that the beryllium concentrations at both urban and non-urban sites were below the detectable limit (0.0008 $\mu g/m^3$).

General urban atmospheric pollution with manganese is occurring as manganese-containing additives are substituted for lead antiknock compounds.[61] In view of the pneumonitis and increased mortality from pneumonia which have in the past been caused by manganese pollution in the vicinity of point sources,[62] it will be important to monitor closely and control within safe limits any future increase in particulate manganese.

In summary, it can be said that of the toxic metals in airborne particles, only lead is present in the urban general atmosphere at average concentrations

which may be hazardous. Other toxic metals (including manganese, mercury, cadmium, arsenic, and beryllium) are potential health hazards in the vicinity of point source emissions.

5.4.1.3 Polycyclic Organic Matter (POM)--These compounds are adsorbed on airborne particles[32] and are included in the "benzene soluble organic fraction". The major human health concern relating to POM is the possible causation of cancer by one or more members of this group of organic compounds. POM is associated with airborne particles emitted from both mobile and stationary sources (such as diesel engines and coke ovens) during the combustion or pyrolysis of hydrocarbons.

A detailed assessment of the health effects of POM has been recently prepared.[63] Occupational studies have clearly demonstrated an increase in lung cancer with exposure to POM. Epidemiological studies in community settings have demonstrated, after correction for smoking, that urban residents have a twofold greater risk for the development of lung cancer as compared to residents of rural environments. It is assumed that POM contributes to some unknown fraction of this increase. It is believed that the risk of lung cancer development by exposure to POM is real, that the magnitude of that risk to the general population cannot be accurately determined based on our current knowledge, that it is almost certainly much less than the lung cancer risk associated with cigarette smoking, but that in real-life situations with concurrent exposures to cigarette smoking and other foreign chemicals, the

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*BaP is the POM for which the most information concerning emissions is available. It is used as an indicator of the presence and concentration of POM as a class. However, the validity of this use of BaP as a surrogate for POM is uncertain.[63]

absolute risk of POM exposure is probably magnified.[63] Urban trend data for a specific POM (benzo(a)pyrene)* show an 80 percent decline from 1966 to 1975.

5.4.1.4 Sulfur Dioxide/Sulfuric Acid/Sulfate Salts--In addition to the survey of the physiological effects of airborne sulfur compounds given in Airborne Particles,[50] two other reviews of this subject have been recently published.[31,64] Therefore, the effects of these substances will be only briefly discussed here. The reader should consult the cited reviews for more detail.

Epidemiological evidence concerning the toxicity of sulfur oxides and sulfates is complicated by the concurrent presence of other particulate matter and other pollutants. However, it is clear that high TSP levels coupled with high sulfur oxides can lead to sickness and fatalities. Observed increases in morbidity and mortality during air pollution episodes are discussed below and in the various reviews cited.

The source of most of the particulate sulfur compounds is fossil fuel combustion, sulfur dioxide being the major form emitted. This gas is quite soluble in water and occurs as sulfurous acid in aqueous airborne particles. It is also oxidized to sulfuric acid, which in turn is neutralized to some extent, forming neutral sulfates.

Much of the evidence suggesting that the sulfate aerosol contributes to the health effects of air pollution is based on experimental work with animals. In various studies at exposure levels of 1000 $\mu g/m^3$ or less, sulfuric acid has not been shown to cause increased pulmonary airflow resistence, chronic changes in liver and lungs, kidney, and pancreas (in combination with other pollutants), decreased carbon monoxide diffusion capacity, and cellular changes in the bronchial mucosa. Because it is a strong acid, sulfuric acid may be expected

to have a greater irritant effect on respiratory mucosa than do sulfate salts. However, because of the presence of ammonia in the respiratory tract, much or all of the inhaled sulfuric acid may be neutralized before contacting the mucosa. The extent to which such neutralization actually occurs will depend on several other factors, including particle size. As noted above, ambient sulfuric acid has been shown to persist even in the presence of more than enough ammonia for complete neutralization.[9]

Animal experiments have shown that neutral sulfate salts are also respiratory irritants to varying degrees, with zinc sulfate and zinc ammonium sulfate being highly irritating and ferric and manganese sulfates being inert. There has been little research on the effect of sulfate salts on humans.

5.4.1.5 Other Particles--A number of other types of particles can be assumed to contribute to the health impact of airborne particles.

Nitric acid vapor and particulate nitrates are formed, to some extent, from nitric oxide in a manner analogous to the formation of sulfuric acid and particulate sulfates from sulfur dioxide. Orel and Seinfeld,[65] in a recent report, compare the formation, sizes, and concentrations of ambient sulfate and nitrate particles. Unlike sulfuric acid, the nitric acid that is formed tends to remain in the gas phase, although it may be an important component of acidic precipitation.[66] Although it is recognized that respirable nitrates may be of health significance, little information is available as to their health effects.

Cigarettes are the source of much of the airborne particle exposures for smokers and of some for non-smokers, especially in indoor environments. Because cigarette smoke particles are small, contain adsorbed pyrolysis products known to be carcinogenic, are deposited and remain for extended periods

in the smokers' lungs, and are deliberately or involuntarily inhaled at high concentrations, they represent a special and extreme case of hazardous airborne particles. The greater susceptibility of smokers to the adverse health effects of airborne particles (e.g., asbestos) and to air pollution episodes is well documented. Although one cannot assume that all of the harmful factors in cigarette smoke are associated with particles, a substantial fraction of them probably is.

5.4.2 Chemical Interactions with Other Pollutants

A recent report examines the role of solid/gas interactions in air pollution.[67] Particle interactions with other air pollutants involve both surface adsorption of pollutant gases and surface-catalyzed chemical reactions. These processes can produce modified particles which may be either more or less hazardous than the original ones.

5.4.2.1 Adsorption--Because many airborne particles incorporate substances which are surface active and because they have large surface areas relative to their mass, these particles are effective sorbents of other air pollutants. Most POM, for example, which is normally emitted as hot vapors, condenses into fine particles upon cooling.[32] Particles can convey into the non-ciliated portion of the lung water-soluble toxic substances that would otherwise have been largely exhaled or removed in the upper respiratory tract[68,69] An analysis of the southern California aerosol on four successive days showed that the most abundant form of carbon in the particles (50 percent) was elemental carbon.[70] Because of their adsorptive efficiency, fine carbon particles can synergistically increase the effects of other pollutants. The co-carcinogenic effect of benzo(a)pyrene has been shown experimentally to be potentiated by the presence of carbon particles with which, in automotive emissions, it is

normally associated.[4] Benzo(a)pyrene[71] and other polycyclic organic matter adsorbed on soot have been shown to be released from the particles by body fluids.

5.4.2.2 Surface Chemical Reactions--Particles participate in various chemical reactions with atmospheric gases. Preeminent, perhaps, are the solution by water droplets of gases and the aggregation of other particles with these water droplets, establishing an aqueous reaction medium where chemical transformations occur. Sulfur dioxide is oxidized to sulfuric acid in the presence of aqueous atmospheric aerosol. The rate of this reaction is dependent on temperature, pH, and the presence of catalytic trace elements. Soluble salts of ferrous iron, manganese, and vanadium are among those which catalyze the oxidation of sulfur dioxide. In a further particle/gas interaction, the sulfuric acid droplets can then be neutralized by atmospheric ammonia.

There are epidemiological and experimental indications that adverse health effects of sulfur dioxide at concentrations normally present in the ambient air are largely or entirely dependent on the coincidental presence of particles.[31] The interaction causing this synergism is presumably due both to the particle-catalyzed oxidation of sulfur dioxide in the environment and to sulfur dioxide oxidation which occurs after dry-particle transport of the adsorbed gas into the warm, humid atmosphere of the respiratory tract. Synergistic effects of sulfur-dioxide and particle exposures are discussed in Airborne Particles.[50] Sulfur dioxide has also been reported to increase synergistically the carcinogenic activity of benzo(a)pyrene particles.[72]

Other aspects of particle surface chemistry are suggested by the report of Linton et al.;[73] for example, in coal fly ash, numerous elements including lead, chromium, manganese, sulfur, iron, potassium, sodium, lithium, and

vanadium have strongly enhanced surface concentrations and leachabilities relative to those of the glassy particle matrix. Lead, for example, while present at a concentration of 0.06 percent on the basis of bulk analysis, had a surface concentration of 4 percent and was highly leachable. Preliminary studies also established the surface predominance of lead, bromine, chlorine, sulfur, phosphorus, potassium, and sodium in automobile exhaust particles. The non-uniformity was attributed to volatilization-condensation processes. Since it is the particle surface which contacts and is extracted by body fluids upon ingestion or inhalation, this non-uniformity will have the effect of increasing the effective dose resulting from inhalation of airborne particles.

5.5 HEALTH COST ANALYSES

With regard to the development of recommended guidelines for the protection of public health, there has been much discussion relating to the desirability and magnitude of safety factors, the desirability of regulating particular types or sizes of particles, the extent to which air quality standards should be designed to protect especially vulnerable groups,* and to the economic benefits to be derived. To provide data related to the latter consideration (cost/benefit trade-offs), a number of studies have attempted to estimate the costs of air pollution. A report by Herman[74] presents a survey of studies published between 1967 and 1977 concerning the health costs of air pollution. It concludes that sulfur oxides and particles are a more serious threat to health than are other

*Such groups might include the aged, the sick, children, those with above-average levels of outdoor activity, and that 10 percent of the healthy population (Airborne Particles) which is reported to exhibit exaggerated responses to sulfur dioxide exposure.[50] The Clean Air Act requires protection of such susceptible groups.

air pollutants. It was stated that "no study surveyed for this report dissents directly or indirectly from the conclusion that sulfur oxides and particulate matter impose the greatest health costs on the American population." A tabular comparison of health cost studies was given in the report by Herman. Those parts of the table summarizing studies which were concerned mainly with sulfur oxide and particle pollution are presented here in Table 13. Estimates from the various studies of the health penalty imposed by these pollutants are given in the eighth row of the table.

Because such studies attempt to assign monetary valuations to essentially indeterminate costs such as lost earnings as well as to more tangible expenses such as medical costs, and because they usually do not include such real but non-quantifiable effects as the psychic and emotional penalties of disease and premature death, they are admittedly only coarse approximations.

5.6 DEFICIENCIES IN THE SCIENTIFIC DATA BASE

Much needed information concerning the effects of airborne particles on public health and welfare is not available. A recent report by the Inhalable Particles Research Committee[75] outlines the most urgent research needs. These include: (1) definitive animal toxicology experiments establishing acute and chronic effects resulting from individual and multiple pollutant exposures not admissible with human subjects, (2) comprehensive clinical studies of physiological responses in human volunteers to low level individual and multiple pollutant exposures, and (3) expanded collection and evaluation of epidemiological data in communities selected so that relationships between particle pollution and observed health effects can be described.

To support the health oriented research there is need for more detailed characterization of particulates - sizes, quantities, composition, and distribution--plus a better understanding of the physical and chemical transformations of particles between their emission and their deposition in lungs and on soils, foilage, etc. Finally, fundamental to all the above research is the refinement of techniques capable of measuring those properties of particulate pollution that emerge as important.

TABLE 13. COMPARISON OF

Reference	72 Lave and Seskin	73 Jaksch and Stoevener	74 Waddell	75 Finklea et al.	76 Johnson et al. (EPA)
Year(s) studied	1963	1969-1970	1970	1970	1970
Pollutant(s)	sulfates particulates	particulates	sulfur dioxide, particulates, sulfates	sulfates	sulfur dioxide, sulfates, particulates
Illness	respiratory disease, cardiovascular disease, and cancer	respiratory diseases, allergies, skin diseases, circulatory, digestive, genitourinary disorders, eye diseases	irritation symptoms, heart and lung (elderly), asthma, acute lower respiratory, chronic bronchitis	heart and lung disease, lower respiratory (LRD) disease (children), chronic respiratory disease (CRD), asthma	Acute resp.--influenza, a. bronchitis, pneumonia Chronic resp.--emphysema, c. bronchitis, bronchiectasis, d. interstitial pneumonia
Death	yes	no	yes	yes	no
Measurement of pollutant(s)	no measurement, 50% improvement assumed	18 Portland monitors; 24-hour readings every 4 days; isopleths drawn, values assigned to people taking account of differ- ent exposures due to home, work, travel	used the average 26% improvement in pollution levels reported for 1970	NASN sulfate readings extrapolated to four types of population density (less than 2500 per city to more than 2M); 1-24 monitors per type in each of 9 regions	annual SMSA averages of each pollutant; 168 SMSAs; center-city NASN readings.
Dose-response function	inferred from previous epidemiological studies	regression analysis: age, sex, marital status, no. in household, race, fitness, drinking, smoking, occupation, humidity, temperature, 1 pollution measure	death: 26% abatement reduces death rate 2.3% (estimated from Lave & Seskin 50%-4.5% relation); illness: reductions estimated from CHESS data	dose-response functions constructed for acid sulfate aerosols from a number of studies, includ- ing CHESS; "best judgment" rather than mathematical fit used	excess disease rates from 9 CHESS study communities applied to SMSAs with same combination and levels of pollutants
Population at risk	nationwide	2,500 Portland residents	irritation-50M, asthma-4M, heart & lung-4M, acute lower-50M, ch. bronch.-6M	heart and lung--3.7M, 4.1M asthmatics, 25.2M children, 137.4M-- premature death	48.8M center-city res.
Results: additional cases of disease or death	50% abatement would cut illness and death from respiratory dis- ease 25%, cancer 15%, and cardiovascular disease 10%	20 micrograms per cubic meter increase to 80 microgram particulate levels had "minimal effect" on cost of outpatient visits	illness: reductions: irritation: 75-100%, heart and lung: 10-30%, others: 10-50%	13,000 excess deaths, 44.7M aggravated heart and lung, 6.8M excess asthma attacks, 0.376M excess LRD, 0.565M nonsmoker excess CRD symptoms	Acute: 1,901,000 cases (adult--17-65); 5,863,000 cases (children) Chronic: 679,000 (adult)
Results: dollar estimate(s)	Savings: respiratory- $1,222M; cancer-$390M; cardiovascular-$468M	additional 3.5¢ per visit or $1000 per year	$0.7-4.4B death:-- $0.9-3.2B illness:-- savings if 26% reduction in air pollution	--	Acute: $475.9 million Chronic: $104.6 million
Definition of estimate	medical costs and lost earnings	medical costs only	illness and death: medical costs and lost earnings	--	medical costs, lost earnings, housewife disability, lost school
Scope of estimate	nationwide	150,000 members of Kaiser-Permanente Medical Care Program in Portland, Oregon	approximately 155M people -urban population nationwide	Nationwide	168 center cities: 48.8 million people

HEALTH COST STUDIES[74]

[77] Liu and Yu	[78] Carpenter et al.	[5] Gregor	[79] Heintz, Hershaft, Horak	[80] Lave and Seskin
1970	1972	1968-1972	1970	1979 (projected)
sulfur dioxide, particulates	sulfur dioxide, particulates	sulfur dioxide, particulates	sulfur dioxide, sulfate compounds, particulates	sulfur oxides, particulates
yes: not specified	respiratory and heart disease	no	same as Waddell	all illness
yes	no	yes	same as Waddell	yes
SO_2: 1968-70 SO_2 averages in 40 SMSAs where average exceeded 25 micrograms per cubic meter; one per SMSA, particulates: 1968-70 averages from same SMSAs	pollutant measurements used to classify neighborhoods according to three levels of SO_2 and particulates[2] in Allegheny County (encompasses Pittsburgh)	sulfur dioxide: 5-year mean of annual averages measured at 5-49 stations; particulates: 5-year mean of annual averages, 42-47 stations; means for each census tract in Allegheny Co. (Pittsburgh)	same as Waddell: no measurement; assumed 26% improvement in pollution levels	smallest SO_2 reading, arithmetic mean of particulate readings: taken biweekly in 1960-1961 and 1969 in each of 112-117 SMSAs
illness-calculated from 162 CHESS observations using "Monte Carlo" method death-regression analysis: % over 65, income, education, 3 weather variables explains 82% of death rate	regression analysis: sex, age, occupation, smoking, neighborhood median income, how hospital bill paid, whether surgery performed, % occupancy of hospital, race, 3 measures of pollution	regression analysis: % adults with high school education, population density, precipitation, temperature, 2 pollution measures	same as Waddell, see "Results" below	regression equation: 2 pollution variables, % of people 65 and over, income, population density, % non-white explains 83% of the death rate
40 SMSAs; app. 64 million (1970 Statistical Abstract)	32,600 patients in 28 hospitals	app. 950,000 Allegheny County residents	same as Waddell	nationwide
SO_2 and particulates account for app. 1% of total death rate SO_2 accounts for 7% of excess illness rate, particulates 28%	133,600 additional hospital-days, plus 1-3.5 days longer in hospital suffered by residents of more polluted neighborhoods	1% SO_2 and particulate reduction in 1968-1972 would have reduced death rate .01-.06/100,000 for younger than 45; .1-.5, 45-64; .3-12.6, 65+	26% abatement reduces death rate 2.33% illness savings see Waddell	EPA-projected 58% and 88% reductions from 1971 levels of particulates and sulfur oxides by 1979 would reduce U.S. death and illness
death-$1,930.6 million illness-$249.5 million	$9.9 million extra hospitalization costs in 1972	potential saving of $7.5 million per year	death-$3.2 billion: illness-$2.5 billion: savings if 26% reduction in air pollution	$16.6 billion savings in 1979 (7% of 1972 U.S. medical costs projected to 1979, discounted at 6%)
death-lost earnings, illness-medical costs, lost earnings, "psychic" costs	medical costs only	"willingness to pay"-$200 for 1/1000 reduction in annual chance of dying	Waddell figures updated to account for rising wages, population increase, etc.	medical costs, lost earnings, and housewife equivalent earnings
approximately 64 million people in 40 SMSAs	32,600 respiratory and heart hospital patients in Allegheny County	Allegheny County (Pittsburgh area) residents	approximately 155 million-urban population nationwide	nationwide

6. EFFECTS OF PARTICULATE MATTER ON PUBLIC WELFARE

The effects of airborne particles on public welfare consist, chiefly, of adverse effects on plants, animals, and soils; on materials; on visibility and other aesthetic conditions; on meteorological conditions; and possibly on climate.

6.1 ECOLOGICAL EFFECTS

Airborne particles can cause injury to vegetation. The type of injury which may occur is dependent on particle size and composition. Excessive deposits of dust particles, such as those from cement plants, can cause crusts to form on the leaves, twigs, and flowers of plants if moisture is present. The presence of the crust prevents sunlight from reaching the leaves, thus inhibiting the process of photosynthesis. Studies of these phenomena have been made in the vicinities of dust-emitting cement kilns and are extensively reviewed in Airborne Particles.[50]

The stomata in the leaves may become clogged if they are on the upper leaf surface. Depending on their chemical composition, dusts can cause direct injury to leaves and flowers. Particles containing fluorides, magnesium oxides, and soot-containing particles have been shown to cause vegetational injury.

Fine particulate matter (ambient aerosol) is the principal conveyer of acid in the atmosphere.[36] The effects of acidic precipitation on ecosystems are briefly reviewed in Airborne Particles[50] and more extensively in the numerous contributions to a Symposium on Acid Precipitation.[76] Acidic precipitation, containing sulfuric, nitric, and hydrochloric acids, can

severely inhibit the growth and health of trees and other plants both by direct contact and by acidification of the soil. The effects on domestic plants can have substantial agricultural[77] as well as natural ecological impact.

Several analyses of the effects of acidic precipitation on specific ecosystems have been presented. For example, Schofield[78] estimated that fish populations have been adversely affected by acidification in approximately 75 percent of the high-elevation Adirondack lakes. Some of these lakes are now so acidic that fish cannot survive.

Toxic material in airborne particles can also injure terrestrial animals. An example is the fluoride poisoning which has been observed in cattle and horses grazing near point sources (such as aluminum processing plants).[79] Lead poisoning of wildlife, domestic animals, and aquatic organisms is reviewed in Air Quality Criteria for Lead.[29]

6.2 EFFECTS ON MATERIALS

The damaging effects of particulate matter on materials are extensively reviewed in Airborne Particles.[50] These effects are due not only to the attack of chemically active components such as sulfuric and nitric acids but also to the action of adsorbed or dissolved gaseous pollutants, including sulfur oxides, nitrogen oxides, hydrogen sulfide, ammonia, and ozone. The effects include corrosion of metals, deterioration and discoloration of paint, erosion of stone and masonry, fading and embrittlement of plastics, fading and weakening of textiles, and peeling of asphalt surfaces. An estimate of 3.8 billion dollars as the annual cost of such damage is cited. This estimate did not include costs associated with surface soiling.

Using an economic model of household behavior, Watson and Jaksch[80] estimated welfare gain from less soiling using empirical estimation of a physical soiling function, a behavioral frequency-of-cleaning function, and total current expenditures for cleaning derived from two earlier studies. A behavioral frequency-of-cleaning function and estimates of total cleaning expenditure were obtained by analyzing data gathered for Philadelphia, including data from a cross section survey of 1090 households. Although the results are limited by the empirical data, this preliminary analysis for 123 SMSA's* found that the benefits of less soiling when the federal primary particulate standard is attained would range from $537 to $3816 million per year in 1971 dollars. When soiling benefits and health and materials benefits (obtained from other studies) were combined and compared with control costs, Watson and Jaksch found tentatively that the secondary annual particulate target (60 micrograms per cubic meter) would provide the largest net benefit.

6.3 ATMOSPHERIC AND CLIMATIC EFFECTS

Absorption and scattering of sunlight by aerosol particles cause decreased visibility and decreased intensity of the sunlight which reaches the ground. It has been estimated that such scattering and absorption are responsible for a radiation loss of 20 percent in parts of the rural midwest, an effect of potentially great agricultural impact. Absorption and reflection of sunlight by the aerosol and by particle-enhanced cloud cover may also be responsible, to some degree, for the recent cooling trends in regional and global climate. Although the significance of this effect on climate is still uncertain, it may prove to be of importance.

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*Standard Metropolitan Statistical Areas.

Airborne particles influence cloud formation and rainfall by serving as condensation nuclei. The effect is well documented in areas downwind from urban and industrial locations. It is estimated that many urban centers cause an increase in downwind precipitation of 5 to 15 percent.

These effects of airborne particles on visibility, insolation, cloud cover, global temperature, and precipitation are discussed and documented in detail in Airborne Particles.[50]

6.4 AESTHETIC EFFECTS

Aesthetic and nuisance effects of airborne particles include such phenomena as the general soiling of clothing, buildings, and other exposed surfaces, the association of unpleasant odors with airborne particles,[50] the obscuration of scenic vistas, the cultural loss incurred by degradation of exposed architectural and historical artifacts, and the diminished opportunities for pleasant outdoor activity, including recreational enjoyment of natural ecosystems.

Although the costs of these effects are not normally quantifiable, it is apparent that they are significant and are a major factor in public assessment of the undesirability of airborne particle pollution.[81] The 1969 criteria document, Air Quality Criteria for Particulate Matter,[81] presented the conclusion that total suspended particulate (TSP) levels above 70 mg/m^3 (annual geometric mean) evoke public awareness and/or concern.

6.5 Trends in Visibility

Recent studies suggest that fine particulate sulfates may be a principal cause of visibility reduction associated with air pollution in areas as diverse as Los Angeles,[82] the eastern U.S.,[83] and the southwest.[84]

Widespread atmospheric hazes observed throughout the eastern U.S. are apparently increasing with regional SO_x emissions. Trijonis and Yuan[83] analyzed visibility/pollution relationships at 12 northeastern sites. They found that visibility is not substantially better in non-urban areas than in metropolition areas of the northeast, averaging only 9 to 12 miles. From the middle 1950's to the early 1970's, visibility exhibited only slight downward trends in large metropolition areas but decreased on the order of 10 to 40 percent at surburban and nonurban locations. Over the same period, visual range declined remarkably during the third calender quarter relative to other seasons, making the summer months the worst season for visibility. Examination of meteorological trends at some sites indicates a slight decrease in maximum daily average temperature and a slight increase in humidity, but these are not sufficient to account for the visibility trends. Additional work is needed to determine whether the visibility changes influenced the climate trends.

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